

Sulfate Production and Migration Beneath Dairy Facilities

New Mexico Environment Department (NMED)



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Remediation Oversight Section
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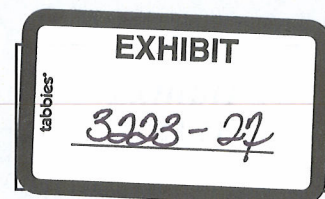


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I. Introduction

The purpose of this review is to illustrate that discharges of dairy wastewater have been shown, and have a high probability to cause, sulfate contamination in ground water above the New Mexico Water Quality Control Commission (WQCC) standard of 600 milligrams per liter (mg/L), thus requiring at a minimum, that ground water samples at dairies be analyzed for sulfate. This review presents the sulfur cycle showing that in New Mexico's subsurface environment the primary fate is the production of sulfate. Sulfate properties are presented indicating that sulfate has a very low potential for retardation in the subsurface and can easily migrate to ground water along a wetting front. Dairy waste contains large amounts of sulfur that readily oxidize to sulfates. It would be remiss of this review to not recognize that naturally occurring sulfates occur throughout New Mexico's aquifers. The review presents general and specific information of New Mexico's major aquifers where the primary numbers of New Mexico dairies are located. It includes a description and evaluation of sulfate concentrations in the Middle and Lower Rio Grande aquifers, the Middle-Lower Pecos aquifer, and the High Plains aquifer, specifically the Ogallala aquifer. Site specific ground water sulfate data is presented that shows ground water downgradient from dairies and a confined animal feeding operation have greater sulfate concentrations than upgradient ground water.

II. Sulfur Cycle

The fate of sulfur is complex and may undergo chemical degradations before it is transported to ground water. The major divisions of the sulfur cycle that are involved with the fate and transport of sulfur are: (a) decomposition of organic sulfur compounds; (b) microbial assimilation or immobilization of simple sulfur compounds and their biocell incorporation; (c) oxidation of inorganic ions and compounds (i.e. sulfides, thiosulfate, polythionates, and elemental sulfur); and (d) reduction of sulfate and other anions to sulfide (Alexander, 1977). Figure 1.0 represents the sulfur cycle.

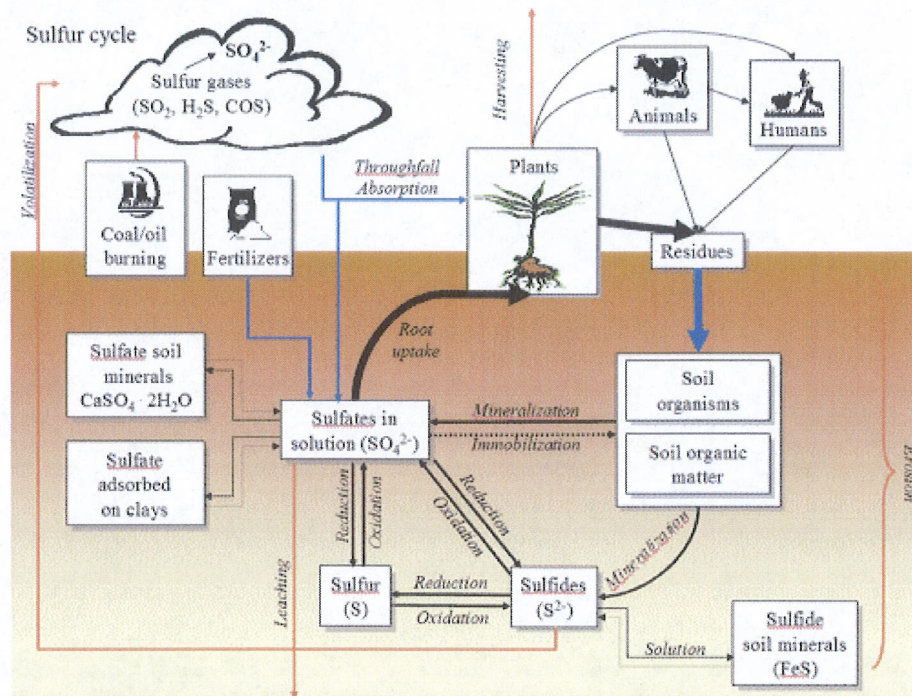
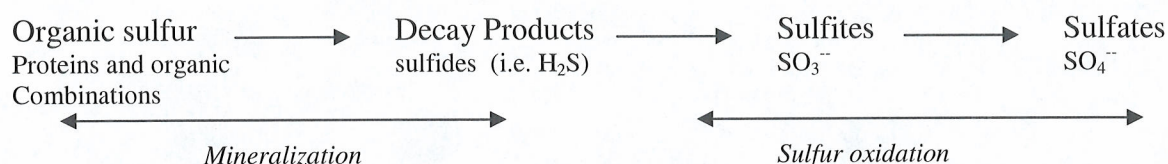
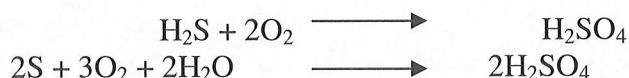


Figure 1 - Sulfur Cycle (UBC – SoilWeb, 2009)

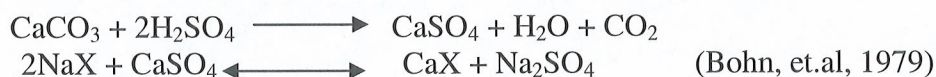
When incorporated in the soil, the proteins of plant and animal tissues are hydrolyzed by the micro flora to the amino acid stage. Microbial activities attack the amino acids and other sulfur containing molecules and organic sulfate and sulfide accumulate. In aerated environments, the combined sulfur is primarily metabolized to sulfate. The transformation of sulfur resembles the bio-conversion of nitrogen. For sulfur the primary sulfide-oxidizing bacteria are of the genus *Thiobacillus*. Reduced sulfur compounds are oxidized to sulfite through the enzyme sulfide oxidase, and the sulfite is further oxidized to sulfate (Chapelle, 2001). Both compounds are largely in organic forms in the soil and mineralization occurs (Alexander, 1977). Mineralization is the overall process of conversion of sulfur from an organic to an inorganic form as a result of microbial decomposition (UBC- SoilWeb, 2009). Once mineralized, these inorganic sulfur compounds are oxidized similarly to the nitrification process. The mineralization process may be characterized as follows (Brady, 1974):



As the cycle shows, organic sulfur can be mineralized to sulfate through oxidation reactions, such as:



Please note that this sulfur oxidation process is an acidifying process and may reduce the alkalinity of soils (Brady, 1974). Furthermore, the reclamation of sodic soils may include sulfur amendments to not only reduce alkalinity but improve soil permeability by exchanging sodium on soil particles, thus producing sodium sulfate, which is capable of leaching. The basic reaction sequence is:



The sulfur cycle does show the reduction of inorganic sulfur compounds to sulfide in aneorobic conditions. This process is either assimilatory or dissimilatory sulfate reduction. Assimilatory reduction occurs when microorganisms are capable of reducing sulfate to sulfide for their metabolic sulfur needs. Dissimilatory sulfate reduction produces hydrogen sulfide in aneorobic conditions, whereby sulfate acts as the electron acceptor in the oxidation of organic carbon or elemental hydrogen (Chapelle, 2001).

“Behavior of sulfur in ground-water systems often reflects the truncation of the cycle due to lack of energy input.” (Chapelle, 2001) Truncation has been observed to occur after sulfide oxidation because of interactions with the iron cycle. An aquifer system that contains pyrite may oxidize the resulting in an increase in sulfate concentrations. If an aquifer system contains a mineral source of sulfate (i.e gypsum) dissolution of that source may cause increases in sulfate concentrations, especially if the aquifer is aerobic (Chapelle, 2001).

Thermodynamic diagrams (Eh/pH) show the stability areas of different species in an aqueous solution. Stability areas are presented as a function of pH and electrochemical potential scales. The system can contain several types of species, such as dissolved ions, condensed oxides, hydroxides, oxides, etc. Usually a large amount of different species exist simultaneously in the aqueous mixtures in fixed Eh-pH-conditions. The Pourbaix diagrams simplify this situation by showing only the predominant species and which content is highest in each stability areas. The lines in the diagrams represent the Eh-pH-conditions where the content of the adjacent species is the same in the equilibrium state (Roine, 2006).

The following sulfur thermodynamic diagram shows the stability areas of different species.

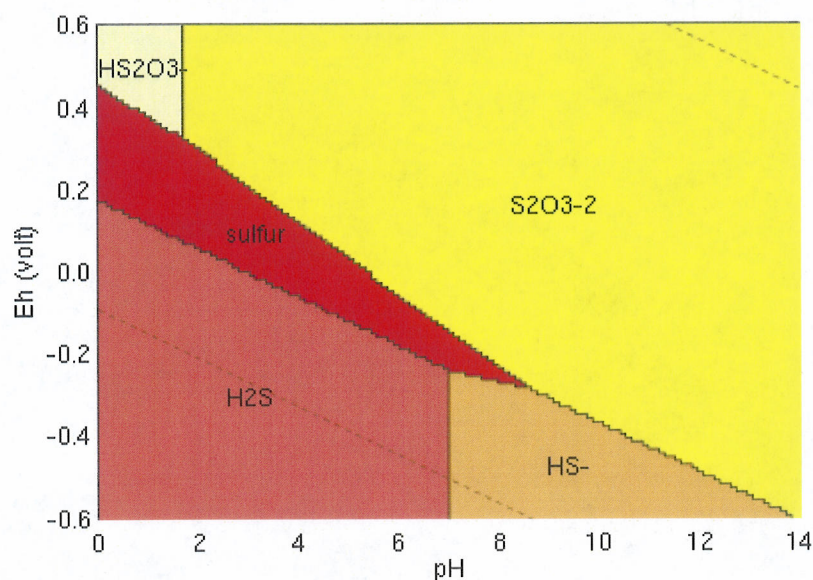


Figure 2 - Partial Eh-pH Diagram of the Sulfur System (Chenoz, 2009)

Generally aquifers in New Mexico have a pH ranging from 6-8 with an Eh value within the -0.2 to +0.2 volt range indicating that the most stable sulfur species are sulfate, elemental sulfur and sulfide. Elemental sulfur is stable only under acid conditions (Bohn, 1979). Typically aerobic aquifers will have sulfate as the most prevalent species while sulfide, as hydrogen sulfide, will be more prevalent in anaerobic aquifer conditions.

III. Sulfate Properties

Understanding the properties of sulfate should be understood for contaminant fate and transport. Since sulfate is a major ion found in soils, it is retained weakly and is rather mobile. Nitrate and chloride move through soils along the wetting front and sulfate and bicarbonate lag behind due to their tendency to form ion pairs, complex ions, or precipitates with soil cations (Bohn, 1979). Sulfate has a minimal sorption that is directly correlated to the soil surface charge. When a soil surface positive charge is low, little to no sulfate sorption has been observed with the changes in sulfate sorbed being dependent on changes in surface positive charge (Marsh, 1987). Sulfate retardation is more dependent on aquifer matrix charge than organic matter. A study conducted

in solute transport at an ash disposal site shows that a sorption model was developed using a retardation factor of 1.1 for sulfate with 1.0 being no retardation (Mudd, 2000). This same study showed that the majority of sulfate leached through the sample following one pore volume leachate. This indicates that there is a minimal amount of sulfate sorbed in the subsurface particles and sulfate will migrate downward to ground water in the vadose zone with the wetting front. Sulfate is mobile in soils and will leach through soils similarly to nitrate (Rehm, 1989).

IV. Sulfur in Dairy Waste

Dairy waste and manure contain sulfur (primarily as organic sulfur) that oxidizes to sulfates and 25 to 30% of total sulfur in manure will be readily available as sulfate (Dairy One Cooperative, 2009). The process of sulfur availability from manure is similar to the process of nitrogen availability. Manure contains sulfur in both organic and inorganic forms. The American Society of Agronomy Crop Science and Soil Science report that sulfur content of dairy solid waste contains 1.5 pounds per ton while liquid manure contains 4.2 pounds per 1,000 gallons (Brown, 2006). The South Carolina Confined Animal Manure Managers Certification Program for dairies report that the sulfur content of fresh manure is 1.2 pounds per ton and that agitated lagoon liquid and sludge contain 2.3 pounds of sulfur per 1,000 gallons (Chastain, 2008). Camberato reports that typical scraped paved lot dairy manure contains 1.7 pounds of sulfur per ton (1996). He further reports that at a field where 24 tons per acre of this type of manure was applied there was 40 pounds per acre of sulfur; 100% more than the recommended application. They further state in an animal manure waste analysis report that the sulfur content was 0.0355% and the agitated sludge contained 2.96 pounds of sulfur per 1,000 gallons. Table 1 below compares these values.

Table 1- Dairy Manure Sulfur Content

Dairy Manure Sulfur Content	Solid Manure (lbs/ton)		Liquid Manure (lbs/1000 gal)	
	Total	Available	Total	Available
Brown, 2006	1.5	0.8	4.2	1.9
South Carolina Certification Program	1.2	NA	2.3	NA
Camberato	1.7	NA	2.96	NA

Approximately 95% of soil sulfur is found in the organic form that, as shown above, is easily converted (mineralized) to sulfate (Rehm, 1989). Dairy wastewater and manure contain primarily organic forms of sulfur. According to the US Department of Agriculture, sulfur mineralization was found to occur ranging from 62 to 127% in an incubation study, indicating sulfate formation (Eghball, 2002). Results from a study evaluating the mineralization of sulfur in two different soils showed that mineralization of sulfur to sulfates occurs rapidly within the first week (Reddy, 2002). If crops in land applications areas do not utilize the sulfate for crop production shortly after sulfate is available, the remaining sulfate may leach to ground water provided a wetting front carries the sulfate downward. Infiltration from wastewater lagoons supplies sulfur to the subsurface that biologically oxidizes to sulfate. This sulfate may be carried to ground water with the wetting front from the leaking lagoons.

Literature review shows that many dairies use copper sulfate as an animal hoof disinfectant and copper and sulfate may be constituents of concern where this practice has been employed. Klingberg states in a report in 2005: "Copper sulfate and other products are used to treat hairy heel warts (considered bacterial caused) and other foot / hoof ailments in dairy cattle. The material is mixed into a foot bath that cows walk through after exiting the milking parlor. Sore feet impact overall animal health and cause dairy cattle to remain less mobile, reducing their desire to go get feed and water, and ultimately reducing milk production." Not only is sulfate identified as a constituent of concern, but copper accumulation may also occur at dairy wastewater discharge points. Moore reports that copper concentrations in manure slurry increased from 4.8 milligrams per liter (mg/l) to 88.6 mg/l (Moore, 2009). New Mexico dairies report that little to no copper sulfate is used.

V. Sulfate in New Mexico's Major Aquifers

Sulfate occurs naturally in many New Mexico's aquifers and site specific evaluations need to be conducted to determine natural and/or anthropogenic sulfate impacts to ground water. Primarily the presence of sulfate in ground water is attributed to the quality of water that enters the aquifer, the type and solubility of minerals present in the subsurface, and the quantity of water lost by evaporation and transpiration (Robson, 1995). The following discussion focuses on the Rio Grande Aquifer (Middle and Lower), the Roswell Basin Aquifer, and the High Plains Aquifer.

1. Rio Grande Aquifer System

The Rio Grande aquifer system consists of a network of hydraulically interconnected aquifers in basin-fill deposits located along the Rio Grande Valley and nearby valleys. The basin-fill aquifers of the system are present in intermountain basins between discontinuous mountain ranges in southern New Mexico and between mountains and tablelands in northern New Mexico. The Rio Grande Rift is the principal geologic feature of the area, which in turn has affected precipitation, runoff, ground water recharge, source material of the basin fill, aquifer characteristics, and water quality. Older basin fill consists of unconsolidated to moderately consolidated lenticular deposits of gravel, sand, and clay interbedded in some areas with andesitic and rhyolitic lava flows, tuffs, and breccias. Younger basin fill consists of unconsolidated, poorly to well-sorted, interbedded Quaternary gravel, sand, silt, and clay. Generally this aquifer system has a small dissolved-solids concentration and contains a preponderance of calcium, bicarbonate, and sulfate ions. This water is classified as a calcium bicarbonate or calcium sulfate type (Robson, 1995). The following figure illustrates the major water types found in the system. Please note that the water types of concern for naturally occurring sulfates are identified as calcium sulfates, sodium sulfate, and magnesium sulfates found primarily within Valencia, Doña Ana and Otero counties.

Figure 62. Ground water in the Rio Grande aquifer system is of varied chemical composition. Generally, calcium bicarbonate or magnesium bicarbonate type water is more prevalent in the northern part of the system, and sodium bicarbonate or sodium sulfate type water is more prevalent in the southern part of the system along the Rio Grande.

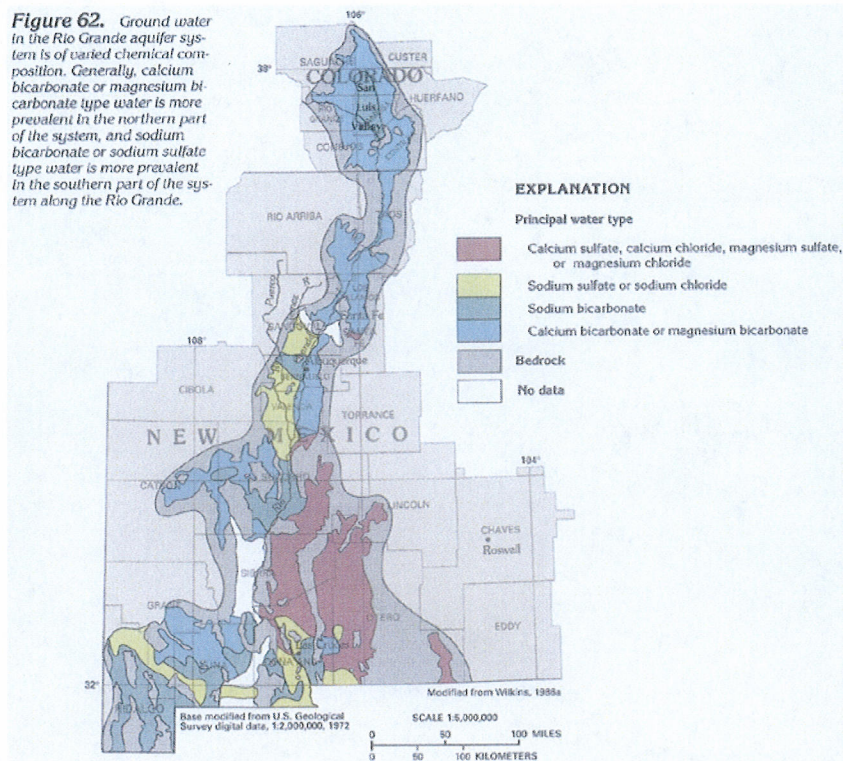


Figure 3 – Principal Water Types of the Rio Grande Aquifer System (Robson, 1995)

Middle Rio Grande

A survey of wells within the Middle Rio Grande Aquifer system indicates a 10 to 90th percentile range of sulfate concentrations between approximately 30 to 700 mg/L with an average of 195 mg/L (Moody, D. 1988).

Lower Rio Grande

A survey of wells within the Lower Rio Grande Aquifer system indicate a 10 to 90th percentile range of sulfate concentrations between approximately 55 to 600 mg/L with an average of 190 mg/L (Moody, D. 1988). A survey of 29 wells in the Mesilla Basin near Las Cruces found sulfate concentrations ranging from 48 to 806 mg/L with an average concentration of 157 mg/L (Bothern, 2003). A study was conducted to determine if salinity contributions to the Lower Rio Grande were from anthropogenic or natural sources (Moore, S., 2008). The study evaluated specific ground and surface water isotopic data along with basic chemical data, including sulfate, that showed that the dominant salinity contributions in the study area were from deep ground water inflow to the Rio Grande, implying that shallower ground water quality was negatively impacted from upwelling deeper aquifer water.

2. Pecos River – Middle Lower

The Pecos River Basin consists of a shallower non-confining alluvial aquifer and an underlying confined carbonate-rock aquifer. Ground water in the carbonate-rock aquifer in the Roswell Basin primarily is present in solution-altered zones in the San Andres Limestone and the overlying Queen and Grayburg Formations. The Grayburg Formation predominantly consists of dolomite and gypsum with interbedded sandstone and shale. The Queen Formation consists of fine-grained sandstone and siltstone with interbedded gypsum (Robson, 1995). The carbonate-

rock aquifer system ranges in thickness from 260 to 600 feet (Welder, 1983). The San Andres and Glorieta Formations of the carbonate-rock aquifer include some evaporite deposits containing gypsum. Dissolution of the evaporite deposits yields sulfate concentrations that generally range from 300 to 1,400 mg/l. Ground water in the western part of the carbonate aquifer in the Roswell Basin generally contains a preponderance of dissolved calcium, magnesium, and sulfate and is classified as either a calcium sulfate or a calcium magnesium sulfate type water (Robson, 1995). The alluvial aquifer primarily consists of the Pecos River flood plain with permeable beds of sand, silts, clay and gravel in the valley fill. The maximum saturated thickness of the shallow aquifer was approximately 250 feet thick at Artesia in 1975 (Welder, 1983). Water in the southern one-half of the alluvial aquifer generally is a calcium sulfate type (Robson, 1995). A study of 55 wells indicates a 10 to 90th percentile range of sulfate concentrations between approximately 400 to 2,500 mg/L with an average of 1,000 mg/L (Moody, D. 1988). The following figure illustrates the major geologic units found in the system.

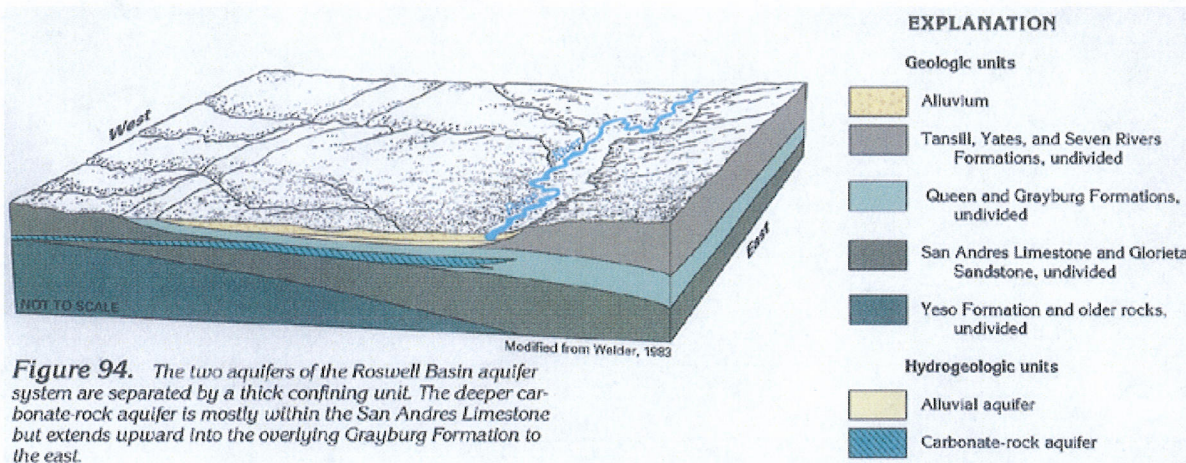


Figure 4 - Geologic Units of the Roswell Basin (Robson, 1995)

NMED conducted a domestic well water fair in October of 2006 to evaluate ground water quality within the southern Chaves County and northern Eddy County area in the vicinity of dairy operations to evaluate if the dairy operations are adversely impacting water quality in the area. Seventy five domestic wells were sampled and 48 of the samples collected showed sulfate results above 600 mg/l, ranging in concentration from 600 to 2,800 mg/l (GWQB, 2006).

3. High Plains Aquifer

The High Plains Aquifer underlies a vast area over many states of approximately 174,000 square miles with parts in eastern New Mexico as shown in Figure 5. The Ogallala Formation is the principal geologic unit in the High Plains aquifer in eastern Colorado and New Mexico. The Ogallala generally consists of an unconsolidated and poorly sorted sequence of gravel, sand, silt, and clay. The Ogallala was deposited by ancient streams that flowed eastward from the Rocky Mountains that deposited random (heterogeneous) sequences of gravel, sand, silt, and clay (Robson, 1995). A subset of data from six wells in the Ogallala in New Mexico collected by the USGS indicate a range of sulfate concentration between 19.1 to 198 mg/L with an average of 95 mg/L (Fahlquist, L. 2001). A larger study of 174 wells indicate a 10 to 90th percentile range of

sulfate concentrations between approximately 20 to 300 mg/L with an average of 85 mg/L (Moody, D. 1988).

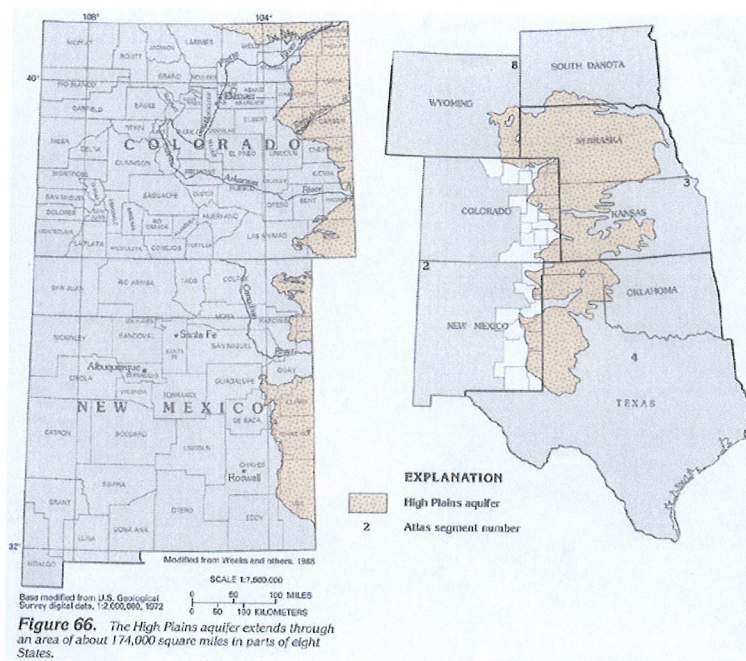


Figure 5 – Extent of High Plains Aquifer (Robson, 1995)

VI. Sulfate at New Mexico Dairies and Animal Feeding Operations

The following discussion presents ground water sulfate concentrations measured at specific dairies throughout New Mexico. Ground water was analyzed for sulfate in a study conducted by NMED around 1985 to determine the effectiveness of manure liners at dairy lagoons in the Lower Rio Grande Aquifer. This study evaluated ground water quality at seven dairies. Four of the dairies were located adjacent to each other near Mesquite, New Mexico. Within the Mesquite dairies, ground water analytical results showed a sulfate concentration ranging from 111 to 1,579 mg/L. The most upgradient monitoring well results for this area showed a sulfate concentration ranging from 431 to 618 mg/L, while the most downgradient monitoring well showed sulfate results ranging from 1,195 to 1,279 mg/L. One of the dairies of this study was located north of Las Cruces and sulfate results from ground water samples collected in on-site monitoring wells ranged from 124 to 1,698 mg/L. The most upgradient well showed a maximum sulfate concentration of 461 mg/L, while a downgradient well showed the maximum concentrations measured on site. Another dairy in this study was located in the far southwest part of Doña Ana County within the Lower Rio Grande Valley. Sulfate results from this dairy ranged from 219 to 1,230 mg/L with the most upgradient well showing a maximum concentration of 697 mg/L and a downgradient well showed the maximum concentrations measured on site (Dye, 1985).

NMED collected ground water samples in 2006 and 2007 from monitoring wells located throughout the seven Mesquite dairies located adjacent to each other in a row. Sulfate results ranged from 163 to 1,140 mg/L. The most northern monitoring well sampled is located downgradient of a dairy land application area and showed a maximum sulfate concentration of 946 mg/L. However, an adjacent center pivot irrigation well, screened at depths of

approximately 300 feet, showed a concentration of 456 mg/L within this same time period. The closest downgradient monitoring well showed a sulfate concentration of 1,140 mg/L. All 13 sulfate results with concentrations above standards were from monitoring wells downgradient of either dairy lagoons or land application areas with the exception of one well. A sample was collected in 2008 from the most upgradient monitoring well that showed a sulfate concentration of 770 mg/L.

A former dairy located southwest of Bernalillo in the Middle Rio Grande Aquifer had ground water samples analyzed for sulfate. This former dairy has two separate aquifers; the Valley Fill and Upper Santa Fe. Both aquifers are contaminated above standards for nitrate as nitrogen. Ground water sulfate results in the Valley Fill aquifer ranged from 42 to 791 mg/L. Only one result was above standards located at a monitoring well that also shows a high concentration of nitrate as nitrogen of 140 mg/L on December 16, 2009. Ground water sulfate results in the Upper Santa Fe aquifer ranged from 38.7 to 1,010 mg/L. This maximum sulfate results was analyzed from a well located within the former corral area (Metric, 2010).

A review of ground water data from other confined feeding animal operations shows that impacts to ground water quality have occurred. An egg production plant formerly operated in southern Bernalillo County. Recent monitoring well results show ground water contaminated with nitrate as nitrogen, total dissolved solids, chloride and sulfate. The most upgradient monitoring well results showed a sulfate concentration of 130 mg/L, while monitoring wells located within the highest contamination showed sulfate results ranging from 780 to 4,100 mg/L (EA Engineering, 2010).

VII. Sulfate and Regulatory Requirements

As defined by the Water Quality Control Commission (WQCC) Regulations, a water contaminant means any substance that could alter if discharged or spilled the physical, chemical, biological or radiological qualities of water. As shown above, dairy waste, either liquid or solid, contains sulfur that can oxidize to form sulfate, a water contaminant. A WQCC numeric standard of 600 mg/L has been established for sulfate. To determine if a discharge may alter the qualities of water, a wastewater sample needs to be analyzed for sulfur and sulfate. To provide protection of ground water and to determine if the sulfate impacts to ground water occur, ground water samples at dairies need to be collected and analyzed.

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